# This Page Is Inserted by IFW Operations and is not a part of the Official Record

### **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

### IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.

#### ORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau

DE



### INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6: C03C 13/06, 13/00

(11) International Publication Number:

WO 96/16913

(43) International Publication Date:

6 June 1996 (06.06.96)

(21) International Application Number:

PCT/EP95/04730

A1

(22) International Filing Date:

30 November 1995 (30.11.95)

(30) Priority Data:

P 44 43 022.1

2 December 1994 (02.12.94)

Published

With international search report.

Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

NO, NZ, PL, SI, SK, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT. SE).

(81) Designated States: AU, BR, CA, CN, CZ, FI, HU, IS, JP, KR.

SAINT-GOBAIN [FR/FR]; Les Miroirs, 18, avenue d'Alsace, F-92400 Courbevoie (FR).

(72) Inventors; and

(75) Inventors/Applicants (for US only): LOHE, Peter [DE/DE]; Ritterstrasse 5, D-67122 Mutterstadt (DE). HOLSTEIN, Wolfgang [DE/DE]; Herderstrasse 2, D-35315 Homberg (DE). SCHWAB, Wolfgang [DE/DE]; Schönauer Strasse 25, D-68723 Plankstadt (DE).

(71) Applicant (for all designated States except US): ISOVER

(74) Agent: KADOR & PARTNER; Corneliusstrasse 15, D-80469 Munich (DE).

(54) Title: A MINERAL FIBER COMPOSITION

(57) Abstract

A biodegradable mineral fiber composition, characterized by the following constituents in percent by weight: SiO<sub>2</sub>: 45 to 55; Al<sub>2</sub>O<sub>3</sub>: 0 to less than 4; Fe<sub>2</sub>O<sub>3</sub>: more than 7 to 15; CaO: 18 to 35; MgO: 5 to 15; Na<sub>2</sub>O+K<sub>2</sub>O: 0 to 10; P<sub>2</sub>O<sub>5</sub>: 0 to 5; impurities: 0 to 2.

### FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

		GB	United Kingdom	MR	Mauritania
AT	Austria	GE	Georgia	MW	Malawi
ΑÜ	Australia		Guinea	NE	Niger
BB	Barbados	GN		NL	Netherlands
BE	Belgium	GR	Greece	NO	Norway
BF	Burkina Faso	HU	Hungary	NZ	New Zealand
ВG	Bulgaria	ΙE	Ireland	PL	Poland
BJ	Benin	IT	Italy	PT	Portugai
BR	Brazil	JP	Japan	RO	Romania
BY	Belarus	KE	Kenya	RU	Russian Federation
CA	Canada	KG	Kyrgystan	SD	Sudan
CF	Central African Republic	KP	Democratic People's Republic	SE	Sweden
CG	Congo		of Korea		Slovenia
CH	Switzerland	KR	Republic of Korea	SI	Slovakia
CI	Côte d'Ivoire	ΚZ	Kazakhstan	SK	
CM	Cameroon	LI	Liechtenstein	SN	Senegal
	China	LK	Sri Lanka	TD	· Chad
CN		LU	Luxembourg	TG	Togo
CS	Czechoslovakia	LV	Larvia	TJ	Tajikistan
CZ	Czech Republic	MC	Monaco	TT	Trinidad and Tobago
DE	Germany	MD	Republic of Moldova	UA	Ukraine
DK	Denmark	MG	Madagascar	US	United States of America
ES	Spain		Mali	UZ	Uzbekistan
FI	Finland	ML		VN	Viet Nam
FR	France	MN	Mongolia		
GA	Gabon			,	

WO 96/16913 PCT/EP95/04730

#### A mineral fiber composition

This invention relates to a mineral fiber composition which is biodegradable, i.e. the fibers decompose as soon as they come in contact with a physiological milieu.

The prior art already describes some mineral fiber compositions which are said to be biodegradable.

Biodegradability of mineral fiber compositions is of great importance since various studies indicate that mineral fibers with very small diameters in the range of under 3 microns are suspected to be carcinogenic, while biodegradable mineral fibers with such dimensions show no carcinogenicity.

However, mineral fiber compositions must also have good workability by known methods for producing mineral wool with a small diameter, in particular the jet process or the external rotary process. This involves in particular a sufficient difference of e.g. 80° between the devitrification and processing temperatures.

The mechanical and thermal properties of mineral fibers, or the products made therefrom, are also of crucial importance. Mineral fibers are used for example for insulating purposes to a great extent. Sufficient temperature resistance of the mineral fibers is necessary in particular for use in the industrial sector.

The problem of the invention is to provide a novel mineral fiber composition which is distinguished by high biodegradability, has sufficient temperature resistance for application in the industrial sector, and can be fiberized well.

The invention is based on the finding that this problem can be solved by a mineral fiber composition which consists substantially of silicon dioxide and alkaline-earth oxides,

and further contains alkali oxides as a melting accelerator and a considerable proportion of iron oxide for increasing temperature resistance.

It has turned out that such mineral fiber compositions fulfill the combination of necessary properties, namely biodegradability, sufficient temperature resistance for insulated objects in industry, as well as good workability in the production of the mineral wool as such and the products. This simultaneously means that the upper devitrification temperature of the melt is preferably under 1300°C.

The subject of the invention is a mineral fiber composition which is biodegradable, characterized by the following constituents in percent by weight:

	•
SiO <sub>2</sub>	45 to 55
Al <sub>2</sub> O <sub>3</sub>	0 to less than 4
Fe <sub>2</sub> O <sub>3</sub>	more than 7 to 15
CaO	18 to 35
MgO	5 to 15
$Na_2O + K_2O$	0 to 10
P <sub>2</sub> O <sub>5</sub>	0 to 5
Impurities	0 to 2

The inventive mineral fiber compositions are readily drawable in particular by the jet process, i.e. one obtains a mineral wool with a low-shot content.

The mineral fibers reach a high temperature resistance of at least 1000°C according to DIN 4102, part 17.

Such mineral fibers show good biodegradability.

The mean fiber diameter is usually 1 to 15 microns, a range of 2.5 to 8 microns being preferred.

The addition of alkali oxides causes a melting point reduction and therefore better workability in the melting process. Furthermore, up to 30% recycled glass can be used advantageously with a sodium-containing mineral wool composition.

The inventive mineral fiber compositions can preferably be melted in melting chambers fueled with fossile fuels, in particular natural gas, at melting temperatures from 1350 to 1450°C. Such melting chambers can produce a homogeneous melt, which is a prerequisite for constant product quality. Homogeneity of the glass melt also facilitates the reproducibility of the fiberizing process and thus of the thermal and mechanical product properties. Furthermore, the constant chemical composition of the thus produced mineral wool leads to controllable biodegradability.

In particular the addition of iron oxide increases the temperature resistance of the mineral wool.

The inventive mineral fiber compositions preferably have the following constituents in percent by weight:

SiO <sub>2</sub>	45 to 53
Al <sub>2</sub> O <sub>3</sub>	0.3 to 3.9
Fe <sub>2</sub> O <sub>3</sub>	more than 7 to 13
CaO	20 to 25
MgO	10 to 15
Na <sub>2</sub> O + K <sub>2</sub> O	3 to 8
Impurities	0 to 2

A content of silicon oxide in the range of 46 to 52% by weight is especially preferred.

With respect to the alkali oxides a range of 3 to 6% by weight is especially preferred. Iron oxide is preferably present in a range between 7,1 and 11% by weight.

To assess biological degradability the standard powder test of the German Glass Society was used. This is an easily conducted method and gives a sufficient measure of biological degradability when used with a simulated physiological lung fluid at 37°C. The method is described in L. Springer, "Laboratoriumsbuch für die Glasindustrie", 3rd edition, 1950, Halle/S: W. Knapp Verlag.

The thermal behavior of the mineral fibers was determined by the so-called "Swedish method". This method uses a silit pipe furnace with a horizontal working pipe open on both sides with a length of 350 mm and an inside diameter of 27 mm. In the center of the furnace there is a ceramic supporting plate with dimensions of 30 x 20 x 3 mm for positioning the test sample. The test sample has dimensions of 12 x 12 mm or 12 mm  $\emptyset$  x 12 mm height. The gross density is normally 100 kg/m<sup>3</sup>. The temperature increase is 5 K/min. The change in test sample height is determined continuously with a reading optic.

The invention will be described more closely in the following using examples.

#### Example 1

A mineral wool was produced with the following composition in percent by weight:

SiO2

47.4

Al<sub>2</sub>O<sub>3</sub>

0.6

Fe <sub>2</sub> O <sub>3</sub>	10.1
CaO	23.5
MgO	10.4
Na <sub>2</sub> O	7.4
K <sub>2</sub> O	0.3
Diverse	0.3

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

An investigation by the modified powder test of the Deutsche Glasgesellschaft yielded a value of 45 mg/kg and thus a value for high biodegradability.

Determination of thermal behavior by the "Swedish method" yielded a temperature resistance of 950°C with 20% height reduction, which can be clearly seen in the corresponding diagram shown by way of example in the single drawing.

#### Example 2

A mineral wool was produced with the following composition in percent by weight:

${\tt SiO_2}$		49
Al <sub>2</sub> O <sub>3</sub>	•	0.3
Fe <sub>2</sub> O <sub>3</sub>		10.0
CaO		23.5
MgO		12

Na <sub>2</sub> O	5.5
Diverse	0.2

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a mean diameter range of 2.5 to 8.0 microns.

An investigation by the modified powder test of the Deutsche Glasgesellschaft yielded a value of 42 mg/kg and thus a value for high biodegradability.

Determination of thermal behavior by the "Swedish method" yielded a temperature resistance of 1000°C with 20% height reduction.

#### Example 3

A mineral wool was produced with the following composition in percent by weight:

SiO <sub>2</sub>	48.7
Al <sub>2</sub> 0 <sub>3</sub>	0.5
Fe <sub>2</sub> O <sub>3</sub> .	10.0
CaO	23.1
MgO	11.9
Na <sub>2</sub> O	5.4
K <sub>2</sub> O	0.1
Diverse	0.3

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into

mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

#### Example 4

A mineral wool was produced with the following composition in percent by weight:

SiO <sub>2</sub>	49.7
Al <sub>2</sub> 0 <sub>3</sub>	0.5
Fe <sub>2</sub> O <sub>3</sub>	9.0
CaO	23.1
MgO	11.9
Na <sub>2</sub> O	5.4
K <sub>2</sub> O	0.1
Diverse ·	0.3

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

#### Example 5

A mineral wool was produced with the following composition in percent by weight:

SiO <sub>2</sub>	50.7
Al 202	0.5

Fe <sub>2</sub> O <sub>3</sub>	8.0
CaO	23.1
MgO	11.9
Na <sub>2</sub> O	5.4
K <sub>2</sub> O	0.1
Diverse	0.3

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

#### Example 6

A mineral wool was produced with the following composition in percent by weight:

SiO <sub>2</sub>	51.7
Al <sub>2</sub> O <sub>3</sub>	0.5
Fe <sub>2</sub> O <sub>3</sub>	7.1
CaO	23.1
MgO	11.9
Na <sub>2</sub> O	5.4
K <sub>2</sub> O	0.1
Diverse	0.2

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

#### Example 7

A mineral wool was produced with the following composition in percent by weight:

SiO <sub>2</sub>		51.7
Al <sub>2</sub> O <sub>3</sub>	₩ -	0.5
Fe <sub>2</sub> O <sub>3</sub>		7.1
CaO	•	25.5
MgO		11.9
Na <sub>2</sub> O		3.0
к20		0.1
Diverse		0.2

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

#### Example 8

A mineral wool was produced with the following composition in percent by weight:

SiO <sub>2</sub>	46.0
Al <sub>2</sub> O <sub>3</sub>	2.5
Fe <sub>2</sub> O <sub>3</sub>	7.1
CaO	27.5
MgO	13.3

PCT/EP95/04730

Na <sub>2</sub> O	3.0
K <sub>2</sub> O	0.1
Diverse	0.5

This composition could be readily fiberized by the jet process at a drawing temperature between 1300 and 1400°C into mineral fibers with a diameter range of 1.0 to 15 microns, a mean diameter range of 2.5 to 8.0 microns being preferred.

#### Claims

1. A mineral fiber composition which is biodegradable, characterized by the following constituents in percent by weight:

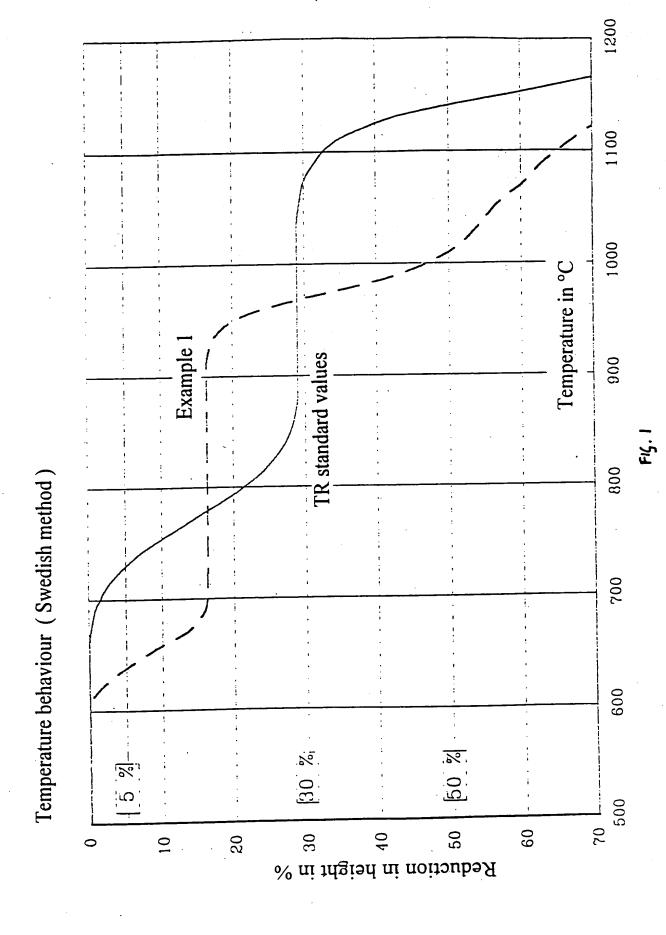
SiO <sub>2</sub>	45 to 55
Al <sub>2</sub> O <sub>3</sub>	0 to less than 4
Fe <sub>2</sub> O <sub>3</sub>	more than 7 to 15
CaO	18 to 35
MgO	5 to 15
$Na_2O + K_2O$	0 to 10
P <sub>2</sub> O <sub>5</sub>	0 <sub>.</sub> to 5
Impurities	0 to 2

2. The mineral fiber composition of claim 1, characterized by the following constituents in percent by weight:

SiO <sub>2</sub>	45 to 53
Al <sub>2</sub> O <sub>3</sub>	0.3 to 3.9
Fe <sub>2</sub> O <sub>3</sub>	more than 7 to 13
CaO	20 to 25
MgO	10 to 15
$Na_2O + K_2O$	3 to 8
Impurities	0 to 2

3. The mineral fiber composition of claim 1 or 2, characterized in that the proportion of silicon dioxide is 46 to 52% by weight.

- 4. The mineral fiber composition of any of claims 1 to 3, characterized in that the alkali oxides are present in a quantity of 3 to 6% by weight.
- 5. The mineral fiber composition of any of claims 1 to 4, characterized in that iron oxide is present in a content between 7 and 11% by weight.



## INTERNATIONAL SEARCH REPORT

Inte: Inal Application No PCT/EP 95/04730

A. CLASS IPC 6	SIFICATION OF SUBJECT MATTER C03C13/06 C03C13/00	
	to International Patent Classification (IPC) or to both national class	affection and IPC
	S SEARCHED	
	documentation searched (classification system followed by classifica-	ation symbols)
Documenta	ation searched other than minimum documentation to the extent that	such documents are included in the fields searched
Electronic	data base consulted during the international search (name of data ba	ase and, where practical, search terms used)
C. DOCU	MENTS CONSIDERED TO BE RELEVANT	
Category *	Citation of document, with indication, where appropriate, of the	relevant passages Relevant to claim No.
Χ .	WO,A,93 22251 (ISOVER SAINT-GOBA November 1993 see page 1, line 37 - page 4, li examples 12,13; tables 1,4	
Х	WO,A,94 14717 (ROCKWOOL INTERNAT 7 July 1994 see page 3, line 3 - line 20; ta	ble 1
A		2-4
Fur	ther documents are listed in the continuation of box C.	X Patent family members are listed in annex.
	ategories of cited documents:	
'A' docum	nent defining the general state of the art which is not dered to be of particular relevance	To later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
which is cited to establish the publication date of another "Y" document of particular relevance: the claimed invention		cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone  'Y' document of particular relevance; the claimed invention  considered to involve an inventive step when the
'O' docum other 'P' docum	nent referring to an oral disclosure, use, exhibition or means nent published prior to the international filing date but	document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.  *& document member of the same patent family
	than the priority date claimed e actual completion of the international search	Date of mailing of the international search report
a	20 March 1996	<b>- 2.</b> 04. 96
Name and	mailing address of the ISA  European Patent Office, P.B. 5818 Patentiaan 2  NL - 2280 HV Rijswijk  Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  Fax: (+31-70) 340-3016	Van Bommel, L

### INTERNATIONAL SEARCH REPORT

information on patent family members

Inter mal Application No
PCT/EP 95/04730

Patent document cited in search report	Publication date	on Patent family member(s)		Publication date
WO-A-9322251	11-11-93	FR-A- AU-B- BR-A- CA-A- CN-A- CZ-A- EP-A- HU-A- JP-T- NO-A- SI-A- SK-A- ZA-A-	2690438 4263293 9305492 2110998 1078708 9302865 0596088 67212 6508600 934725 9300218 146893 9302874	29-10-93 29-11-93 11-10-94 11-11-93 24-11-93 19-10-94 11-05-94 28-03-95 29-09-94 20-12-93 31-12-93 09-11-94 01-06-94
WO-A-9414717	07-07-94	AU-B- CA-A- EP-A- PL-A-	5831694 2152920 0677026 309641	19-07-94 07-07-94 18-10-95 30-10-95